Complete Planarization of via Holes with Aluminum by Selective and Nonselective Chemical Vapor Deposition

著者	坪内 和夫
journal or publication title	Applied Physics Letters
volume	57
number	12
page range	1221-1223
year	1990
URL	http://hdl.handle.net/10097/47682

doi: 10.1063/1.103490

Complete planarization of via holes with aluminum by selective and nonselective chemical vapor deposition

Kazuo Tsubouchi, Kazuya Masu, Nobuyuki Shigeeda, Tatsuya Matano, Yohei Hiura, and Nobuo Mikoshiba[a)]
Research Institute of Electrical Communication, Tohoku University, Katahira 2-1-1, Aoba-ku, Sendai 980, Japan

(Received 12 April 1990; accepted for publication 3 July 1990)

We have developed a new controllable method of selective and nonselective deposition of high quality aluminum by low-pressure chemical vapor deposition using dimethylaluminum hydride with hydrogen. At first, silicon dioxide via holes on silicon substrate were selectively filled with aluminum by thermal decomposition. Then, adding the plasma excitation for 1 min, the aluminum film began to deposit nonselectively on the silicon dioxide as well as the selectively deposited aluminum. Silicon dioxide via holes were completely planarized by the selective and nonselective deposition. The single-crystal structure of aluminum deposited selectively on silicon was observed with a new scanning microreflection high-energy electron diffraction microscope.

Multilevel interconnections are required in the fabrication of very large scale integrated (VLSI) circuits. Aluminum and aluminum alloy are still choice interconnect materials. For the past few years, the chemical vapor deposition (CVD) of aluminum has been investigated for its capability of achieving conformal step coverage,[1–3] selective growth onto Si surface,[4–7] and single-crystal growth on Si wafer.[8] However, the CVD technology is not yet suitable for practical use, because it cannot give the full controllability for selective and nonselective deposition of high quality aluminum. This letter presents, for the first time, a new controllable method of selective and nonselective growth of high quality aluminum on (100) Si and (111) Si versus silicon dioxide (SiO_2) by low-pressure chemical vapor deposition (LPCVD) using dimethylaluminum hydride with hydrogen. We observed the single-crystal structure of aluminum deposited selectively on Si in microregion with a new scanning microreflection high-energy electron diffraction (μ-RHEED) microscope.

The experimental apparatus for the work used here was similar to that described previously except the gas source.[7] The dimethylaluminum hydride [DMAH: $(CH_3)_2AlH$, UBE Corp., Japan] used was a clear viscous liquid and had a vapor pressure of 2 Torr (20 °C), which was ten times higher than that of triisobuthylaluminum. DMAH with carrier H_2 gas was introduced to a horn-type quartz reactor. A 4-in.-diam Si wafer was placed on a heated susceptor. Plain Si wafers and SiO_2-patterned Si wafers were chemically cleaned by H_2SO_4:H_2O_2(4:1) solution with intermediate rinse in de-ionized water. The wafers were then pretreated just before aluminum deposition by a short dip in HF:H_2O (1:25) solution in order to remove residual oxide on Si surface, followed by a 10 min rinse in de-ionized water. Aluminum films were selectively deposited on Si by thermal decomposition of DMAH with H_2. The rf (13.56 MHz) excited plasma was generated prior to the nonselective deposition of aluminum onto the SiO_2 surface.

Aluminum films were deposited at a total pressure of 1.2–3.0 Torr and DMAH partial pressure of $(3–7)\times 10^{-3}$ Torr. The deposition rate (DR) on Si was found to have a surface reaction limited form: $DR = DR_0 \exp(-E/kT)$, where $E = 0.52$ eV in the temperature range of 230–350 °C. The typical deposition temperature was 270 °C and the typical deposition rate was about 800 Å/min selectively into via holes on Si and nonselectively on SiO_2. The electron spectroscopy for chemical analysis revealed that carbon and oxygen were absent from the film. The resistivity of the aluminum film was 3 $\mu\Omega$ cm, which was very close to the bulk resistivity of 2.65 $\mu\Omega$ cm.

A 1-μm-thick aluminum film with smooth surface morphology was deposited on (111) Si as shown in Fig. 1(a). Figure 1(b) shows the reflection high-energy electron diffraction (RHEED) pattern of the aluminum film deposited on (111) Si. Since the single-crystal spot pattern was observed, (100) aluminum was epitaxially grown on (111) Si.

We have found that the aluminum film was not deposited onto thermally oxidized SiO_2 surface, i.e., the aluminum was deposited selectively on Si surface. Via holes of 1 μm in depth and 0.8 μm in diameter were selectively filled with aluminum. Figures 2(a) and 2(b) show selectively deposited aluminum into SiO_2 via holes on (111) Si and (100) Si, respectively. The aluminum of Figs. 2(a) and 2(b) was selectively overgrown after filling the via holes. The overgrown aluminum exhibited a pyramidal shape for the via holes on (111) Si and triangular terrace for those on (100) Si. By determining their own shapes of the single-crystal planes of aluminum, the single-crystal (100) and (111) aluminum were selectively deposited on (111) Si and (100) Si, respectively. In Fig. 2(a), the sidewall triangle of the pyramid was a equilateral one, and the interior angle of the sidewall triangle looking from front view and in parallel to the SiO_2 surface was found to be 55°. Conse-

[a)]Present address: Hewlett-Packard Laboratories Japan, Tokyo, Japan.

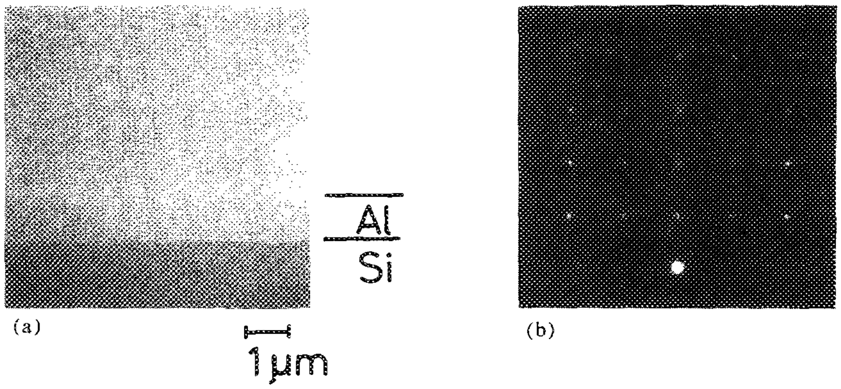

FIG. 1. (a) Secondary-electron microscope photograph of aluminum on (111) Si. (b) Relection high-energy electron diffraction pattern of aluminum film on (111) Si. The spot pattern shows that the aluminum film on (111) Si was (100) single crystal.

quently, the triangular plane of the pyramid on (111) Si was confirmed to be (111) plane of aluminum. The selectively deposited aluminum was also confirmed to be single crystal with a scanning μ-RHEED microscope.

In the conventional RHEED pattern observation, the electron beam was larger than 100 μm Φ, and one could not evaluate the crystal structure in the microregion. The authors have already developed a scanning μ-RHEED microscope in order to evaluate the micrograin structure.[9,10] Three diffraction spots of a RHEED pattern were simultaneously selected for imaging. The scanning μ-RHEED images were obtained from the intensity change of the diffraction spots. We observed the distribution of normally rotated micrograins inside the same plane grain parallel to the surface. Figure 3 shows the scanning μ-RHEED images of the aluminum deposited selectively on (100) Si. The selectively deposited aluminum of Fig. 3 was strongly oriented to (111) from the conventional x-ray diffraction measurement. The scanning μ-RHEED images of Figs. 3(a) and 3(b) were obtained using the intensity change of diffraction spots 333 and 531, respectively. If the crystal plane perpendicular to the surface was rotated inside a (111) grain, the different nonuniform μ-RHEED images should be observed in Figs. 3(a) and 3(b). However, the nonuniform structure in the aluminum lines was not observed, so that the single-crystal (111) aluminum was selectively deposited on (100) Si. The scanning μ-RHEED images of the selectively deposited aluminum on (111) Si also showed that single-crystal (100) aluminum was selectively deposited.

We found that the aluminum was deposited on electrically conductive materials such as p-type Si, n-type Si, and TiN. On the other hand, the aluminum was not deposited on insulating materials such as thermally oxidized SiO_2 and borophosphosilicate glass (BPSG).

In this work, we used the plasma excitation technique when changing the deposition mode into the nonselective one. Figure 4 shows the completely planarized via hole filled with aluminum. At first, the aluminum was deposited selectively on the (100) Si surface by thermal decomposition. Just after the via hole filling, the rf-excited plasma was generated in a quartz reactor. The plasma was con-

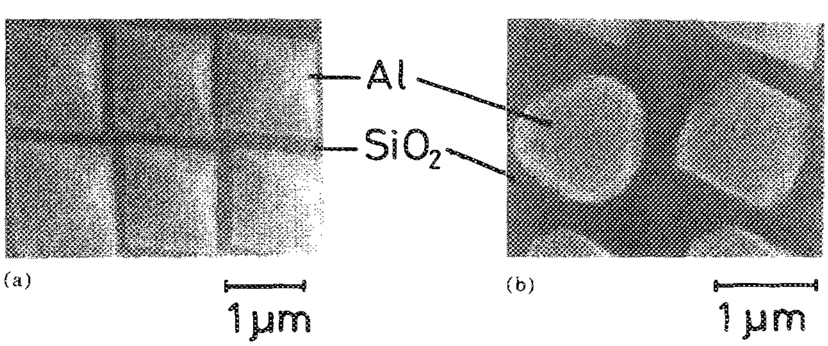

FIG. 2. Secondary-electron microscope photographs of aluminum. (a) Selectively deposited aluminum into silicon dioxide via holes on (111) Si, and (b) via holes on (100) Si. The diameter and depth of the via holes are 0.8 and 1 μm, respectively.

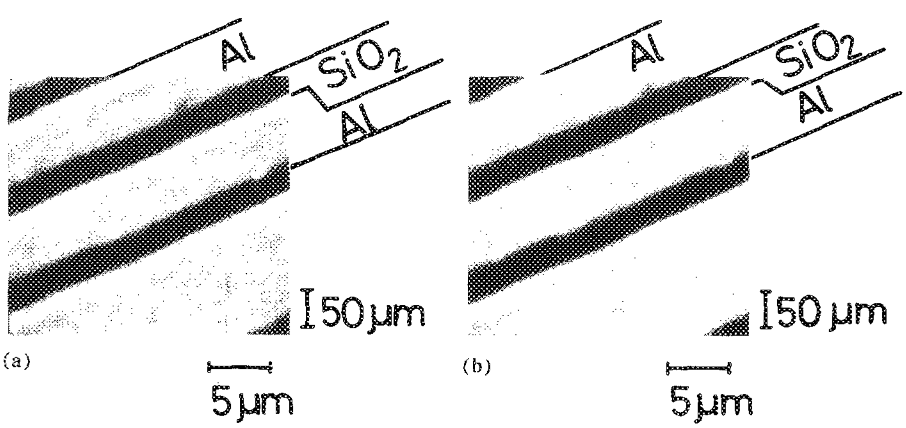

FIG. 3. Scanning μ-RHEED images of aluminum lines on (100) Si. Detection spots were 333 in (a) and 531 in (b). No micrograin structure was observed in the aluminum lines, and the aluminum was confirmed to be (111) single crystal.

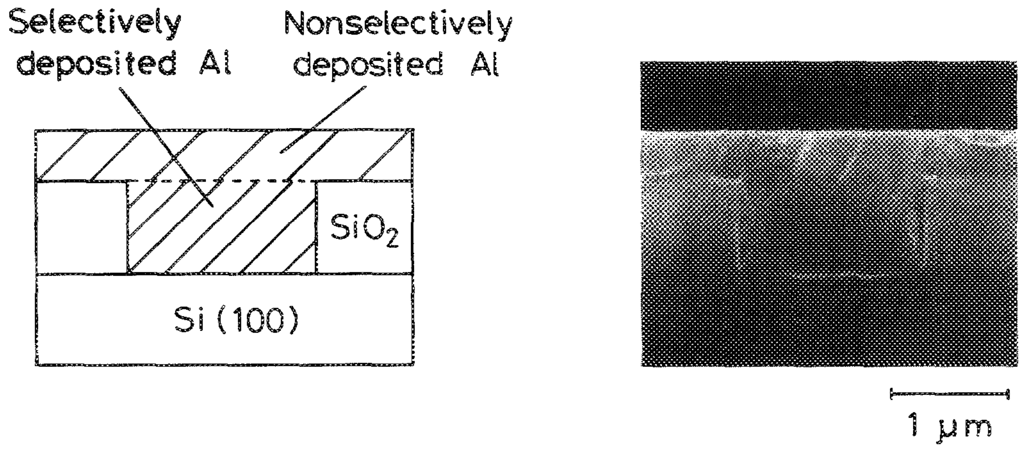

FIG. 4. Complete planarization of via hole filled with aluminum.

fined in a three-separated electrode,[7] where the center ring electrode was powered with the rf power supply and the other two ring electrodes were grounded. In order to avoid charged particle damage onto the wafer, the plasma did not reach the wafer surface. Then, adding the plasma excitation for 1 min, the aluminum film began to deposit nonselectively on the SiO_2 surface as well as the selectively deposited aluminum. After the above 1 min, the nonselective deposition was continued without the plasma. The SiO_2 via hole was completely planarized by the selective and the subsequent nonselective deposition as shown in Fig. 4. In order to deposit aluminum nonselectively, the plasma power density should be larger than 0.04 W/cm^3 and the duration should be longer than 10 s. By RHEED pattern observation, the aluminum deposited nonselectively on SiO_2 was (111) oriented polycrystal.

No hillock formation was observed for aluminum on Si and SiO_2 after annealing at 450 °C for 30 min in N_2 ambient. Erosion at the interface between the selectively deposited aluminum and Si was not observed after the annealing.

In conclusion, we have successfully developed a new controllable method of selective and nonselective deposition of aluminum by low-pressure chemical vapor deposition using dimethylaluminum hydride with hydrogen. The selectively deposited aluminum on silicon surface was confirmed to be single crystal by microregion observation with a new scanning μ-RHEED microscope. Complete planarization of via holes filled with aluminum has the potential for being used in VLSI.

This work was partly done in the Super Cleanroom for Laboratory for Microelectronics, Research Institute of Electrical Communication, Tohoku University.

[1] T. Ito, T. Sugii, and T. Nakamura, *1982 Symposium on VLSI Technology, Digest of Technical Papers, Oiso 1982* (Publication Office, Business Center for Academic Societies, Japan, Tokyo, 1982), p. 20.

[2] M. J. Cooke, R. A. Heinecke, R. C. Stern, and J. W. C. Maes, Solid State Technol. **25** (12), 62 (1982).

[3] R. A. Levy, M. L. Green, and P. K. Gallagher, J. Electrochem. Soc. **131**, 2175 (1984).

[4] T. Amazawa and H. Nakamura, *Extended Abstracts of the 18th (1986 International) Conference on Solid State Devices and Materials, Tokyo 1986* (Publication Office, Business Center for Academic Societies Japan, Tokyo, 1986), p. 755.

[5] T. Amazawa, H. Nakamura, and Y. Arita, *Technical Digest of 1988 IEEE International Electron Devices Meeting, San Francisco 1988* (IEEE, NJ, 1988), p. 442.

[6] C. Sasaoka, K. Mori, Y. Kato, and A. Usui, Appl. Phys. Lett. **55**, 741 (1989).

[7] K. Masu, K. Tsubouchi, N. Shigeeda, T. Matano, and N. Mikoshiba, Appl. Phys. Lett. **56**, 1543 (1990).

[8] T. Kobayashi, A. Sekiguchi, N. Hosokawa, and T. Asamaki, Jpn. J. Appl. Phys. **27**, L1775 (1988).

[9] K. Tsubouchi, K. Masu, M. Tanaka, Y. Hiura, T. Ohmi, N. Mikoshiba, S. Hayashi, T. Marui, A. Teramoto, T. Kajikawa, and H. Soejima, *Extended Abstracts of the 21st Conference on Solid State Devices and Materials, Tokyo 1989* (Publication Office, Business Center for Academic Societies, Japan, Tokyo, 1989), p. 217.

[10] K. Tsubouchi, K. Masu, M. Tanaka, Y. Hiura, T. Ohmi, N. Mikoshiba, S. Hayashi, T. Marui, A. Teramoto, T. Kajikawa, and H. Soejima, Jpn. J. Appl. Phys. **28**, L2075 (1989).